

# A Molecular Matter-Wave Amplifier

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We describe a matter-wave amplifier for vibrational ground state molecules, which uses a Feshbach resonance to first form quasi-bound molecules starting from an atomic Bose-Einstein condensate. The quasi-bound molecules are then driven into their stable vibrational ground state via a two-photon Raman transition inside an optical cavity. The transition from the quasi-bound state to the electronically excited state is driven by a classical field. Amplification of ground state molecules is then achieved by using a strongly damped cavity mode for the transition from the electronically excited molecules to the molecular ground state.

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Recent progress in the application of Feshbach resonances to ultracold atomic gases has led to the production of macroscopic numbers of ultracold molecular dimers starting from either a Bose-Einstein condensate (BEC) [1] or a degenerate Fermi gas [2]. This work has culminated in the production of molecular Bose-Einstein condensates starting from a Fermi gas of  $^{40}\text{K}$  [3] and  $^6\text{Li}$  [4]. The ability to coherently produce a macroscopic number of quantum degenerate molecules opens up new avenues of research in the field of matter-wave optics such as molecular interferometry, the molecular laser, and nonlinear wave mixing between atomic and molecular fields. In fact, the effective Hamiltonians that describe the coherent atom-molecule conversion are formally the same as second harmonic generation in nonlinear optics with a  $\chi^{(2)}$  nonlinear susceptibility.

A major stumbling block on the road to coherent molecular optics is the short lifetime of the molecules formed via Feshbach resonances. While these molecules are translationally very cold, they are vibrationally hot and can decay to lower lying vibrational states via inelastic collisions with other atoms and molecules. For an atomic BEC, the molecules are expected to decay in a time  $\sim 10 - 100\mu\text{s}$  for typical atomic densities [5]. For atomic Fermi gases, recent results indicate that the molecular lifetime can be several orders of magnitude longer due to the underlying Pauli blocking between atoms [6].

Still it would be desirable to create molecules in their vibrational ground state so as to maximize the amount of time with which experiments can be done with them. This can in principle be done using two-photon stimulated Raman photoassociation [7]. This is a relatively inefficient process due to the very small Frank-Condon factors between the free atom pairs and the molecules. This is a consequence of the small spatial overlap between the tightly bound molecular wave function and the large average separation between pairs of atoms in these dilute gases. One way to overcome this difficulty is to use a Feshbach resonance to form weakly bound molecules, followed by a two-photon Raman transition to transfer the

molecules to a low lying vibrational state [8]. The advantages of this procedure are two-fold. First, recent experiments with Feshbach resonances in quantum degenerate gases have demonstrated the ability to readily convert a significant fraction of the atoms into molecules. Secondly, the spatial overlap between the resulting weakly bound states and the ground-state molecules is substantially larger than for that of the free atom pairs, which results in larger two-photon transition rates. Indeed, several experiments using single-photon photoassociation spectroscopy have observed a strong enhancement of the photoassociation rate in the presence of a Feshbach resonance [9].

In this paper we use the idea of Feshbach stimulated photoproduction [8] as the basis for a matter-wave amplifier for ground-state molecules. Active elements such as matter-wave amplifiers are an essential element for the field of matter-wave optics and have previously been demonstrated experimentally for atoms [10].

In our scheme, pairs of atoms in a BEC are coupled to a quasi-bound state of a closed collisional channel via a Feshbach resonance [11]. A two-photon transition couples the quasi-bound molecular state to the vibrational ground state of the molecules. A large amplitude classical electromagnetic field drives the quasi-bound state to an electronically excited molecular state, followed by the emission of a photon into a single mode of a quantized optical cavity field. This second step takes the molecule from the excited state to its vibrational and electronic ground state. By using a strongly damped cavity one can achieve continuous unidirectional coherent amplification of an initial molecular signal field that persists until the atomic condensate is depleted [12]. Although the number of photons in the cavity field is less than one at all times, strong coupling of the molecules to the cavity field can be achieved by controlling the volume and geometry of the cavity as was recently demonstrated in the optical regime with the single atom laser [13].

The discrete mode structure of the cavity allows one to select a particular vibrational state of the electronic ground-state molecules provided the cavity linewidth,  $\kappa$ ,

is less than the vibrational level spacing,  $\delta\nu \sim 1\text{GHz}$ . This is a distinct advantage over using free space spontaneous emission to initially populate the ground state, as in atomic matter-wave amplifiers [10], since spontaneous emission would populate a large number of vibrational levels of the ground state as well as leading to dissociation back into the continuum [14]. As a result, a molecular matter-wave amplifier relying on free space spontaneous emission would require that the initial signal field that is to be amplified be very large in order that stimulated scattering dominates over spontaneous decay into other bound vibrational and continuum states.

Atoms in the BEC are assumed to all occupy the same hyperfine state denoted by  $|0\rangle$ . Pairs of atoms in  $|0\rangle$  are coupled to the quasi-bound molecular state  $|1\rangle$  via a Feshbach resonance with coupling strength  $\alpha$  and with an energy difference between pairs of zero energy atoms and the quasi-bound state given by the difference in the Zeeman interaction energies for the two states,  $\omega_1$  [11]. State  $|1\rangle$  is coupled to an electronically excited molecular state,  $|2\rangle$ , via a classical optical field with Rabi frequency  $\Omega_l$  and frequency  $\omega_l$ . State  $|2\rangle$  is also coupled via a single mode of the cavity field with vacuum Rabi frequency  $g$  and frequency  $\omega_c$  to molecules in their electronic and vibrational ground state,  $|3\rangle$ . The internal energies of states  $|2\rangle$  and  $|3\rangle$  relative to pairs of atoms in  $|0\rangle$  are  $\omega_2$  and  $\omega_3$ , respectively. We assume that  $\Delta = (\omega_2 - \omega_1) - \omega_l \approx (\omega_2 - \omega_3) - \omega_c \gg |g|, |\Omega_l|, \gamma_e$  where  $\gamma_e^{-1}$  is the lifetime of  $|2\rangle$ . Under these conditions the excited state can be adiabatically eliminated, leading to two-photon Raman transitions between  $|1\rangle$  and  $|3\rangle$  with the two-photon Rabi frequency  $\chi(x) = gu(x)\Omega_l^*(x)/\Delta$  where  $u(x)$  is the mode function of the cavity field.

We assume that the atoms and the weakly bound molecules in state  $|1\rangle$  are confined by a trapping potential inside the optical cavity. The state to be amplified,  $|3\rangle$ , may also be confined by a trapping potential or molecules in that state may pass through the cavity as a wave packet [12]. Here we only treat explicitly the case of molecules in  $|3\rangle$  being in a stationary state of their center-of-mass motion. At zero temperature we can use a single mode approximation for the atomic and molecular fields with the center-of-mass wave functions  $\phi_n(x)$  and energies  $\epsilon_n$  for  $|n = 0, 1, 2, 3\rangle$ . The Hamiltonian, in a rotating frame in which the energy of  $|3\rangle$  is given by the two-photon detuning,  $\delta = (\omega_3 + \epsilon_3) - (\omega_1 + \epsilon_1) - (\omega_l - \omega_c)$ , consists of three terms,  $H = \delta \hat{b}_3^\dagger \hat{b}_3 + H_{13} + H_{01}$ , where

$$H_{13} = \frac{1}{2} \chi' \hat{b}_1^\dagger \hat{b}_3 \hat{a} + h.c., H_{01} = \alpha' \hat{b}_1^\dagger \hat{b}_0 \hat{b}_0 e^{i\omega_1' t} + h.c..$$

Here,  $H_{13}$  represents the two-photon transitions between  $|1\rangle$  and  $|3\rangle$  with  $\chi' = -\int d^3x \phi_1^*(x) \chi(x) \phi_3(x)$  while  $H_{01}$  represents the Feshbach resonance coupling of states  $|0\rangle$  and  $|1\rangle$  with  $\alpha' = \alpha \int d^3x \phi_1(x)^* \phi_0^2(x)$  and  $\omega_1' = \omega_1 + (\epsilon_1 - 2\epsilon_0)$ . The operators  $\hat{b}_i$  are bosonic annihilation operators for atoms or molecules in state  $|i\rangle$  and  $\hat{a}$  is the

annihilation operator for photons in the cavity field.

The cavity field is damped at a rate  $\kappa$  with the damping being described by the Born-Markov master equation for a damped harmonic oscillator. The time evolution of the total density operator,  $w(t)$ , is then given by  $dw/dt = -i[H, w] + \kappa (2\hat{a}w\hat{a}^\dagger - \hat{a}^\dagger\hat{a}w - w\hat{a}^\dagger\hat{a})$ . In the bad cavity limit where  $\kappa \gg |\chi'|/\sqrt{N_0}, |\delta|$ , where  $N_0$  is the maximum number of molecules in  $|1\rangle$  or  $|3\rangle$ , the cavity field can be adiabatically eliminated to give a master equation for the reduced density operator,  $\rho = Tr_{\text{cavity}}[w]$ , [15]

$$\dot{\rho} = -i[H_{01} + \delta \hat{b}_3^\dagger \hat{b}_3, \rho] + \kappa |\beta|^2 \left( \hat{b}_1 \hat{b}_1^\dagger \rho \hat{b}_3 \hat{b}_1^\dagger - \hat{b}_3 \hat{b}_1^\dagger \hat{b}_1 \hat{b}_3 \rho + h.c. \right) \quad (1)$$

and  $\beta = \chi'/2\kappa \ll 1$ . The term proportional to  $\kappa|\beta|^2$  in Eq. (1) represents the amplification of molecules in  $|3\rangle$ . Only the  $|1\rangle \rightarrow |3\rangle$  transition occurs because the photons emitted into the cavity are lost before they can be reabsorbed. The first term in Eq.(1),  $[H_{01}, \rho]$ , represents the oscillatory conversion of pairs of atoms into molecules. For  $\kappa|\beta|^2 \gtrsim \alpha'$  the coherent oscillations between atom pairs and quasi-bound molecules are suppressed and the conversion of atoms into molecules dominates over the reverse process.

Equation (1) can be used to derive the equations of motion for the expectation values of the amplitudes,  $\langle \hat{b}_i \rangle = Tr[\hat{b}_i \rho]$ , and the populations,  $\langle \hat{n}_i \rangle = Tr[\hat{n}_i \rho]$  where  $\hat{n}_i = \hat{b}_i^\dagger \hat{b}_i$ ,

$$\begin{aligned} \dot{\langle \hat{n}_3 \rangle} &= 2\kappa|\beta|^2 \langle (1 + \hat{n}_3) \hat{n}_1 \rangle \\ \dot{\langle \hat{b}_1 \rangle} &= -\kappa|\beta|^2 \langle (1 + \hat{n}_3) \hat{b}_1 \rangle - i\alpha' \langle \hat{b}_0 \hat{b}_0 \rangle e^{i\omega_1' t} \\ \dot{\langle \hat{n}_1 \rangle} &= -2\kappa|\beta|^2 \langle (1 + \hat{n}_3) \hat{n}_1 \rangle - \alpha' (i \langle \hat{b}_0 \hat{b}_0 \hat{b}_1^\dagger \rangle e^{i\omega_1' t} + c.c.) \\ \dot{\langle \hat{b}_0 \hat{b}_0 \rangle} &= -2i\alpha' \langle (1 + 2\hat{n}_0) \hat{b}_1 \rangle e^{-i\omega_1' t} \end{aligned}$$

and  $\dot{\langle \hat{b}_3 \rangle} = -i\delta \langle \hat{b}_3 \rangle + \kappa|\beta|^2 \langle \hat{n}_1 \hat{b}_3 \rangle$ . Note that it is the pairing field,  $\langle \hat{b}_0 \hat{b}_0 \rangle$ , rather than the atomic field,  $\langle \hat{b}_0 \rangle$ , that drives the formation of molecules in  $|1\rangle$ . The atomic population is determined by conservation of total particle number,  $d(\langle \hat{n}_0 \rangle + 2\langle \hat{n}_1 \rangle + 2\langle \hat{n}_3 \rangle)/dt = 0$ .

These equations do not form a closed set since the expectation values of the amplitudes and populations couple to the various cross correlations between the modes such as  $\langle \hat{n}_1 \hat{n}_3 \rangle$  or  $\langle \hat{b}_0 \hat{b}_0 \hat{b}_1^\dagger \rangle$ . The simplest approximation is to assume that the correlations factorize at all times, resulting in the closed set of differential equations,

$$\dot{n}_3 = 2(1 + n_3)n_1 \quad (2)$$

$$\dot{b}_1 = -[(1 + n_3) + \bar{\Gamma}/2]b_1 - i\bar{\alpha}P_0 e^{i\bar{\omega}_1 \tau} \quad (3)$$

$$\dot{n}_1 = -2[(1 + n_3) + \bar{\Gamma}/2]n_1 - \bar{\alpha}(iP_0 b_1^* e^{i\bar{\omega}_1 \tau} + c.c.) \quad (4)$$

$$\dot{P}_0 = -2i\bar{\alpha}(1 + 2n_0)b_1 e^{-i\bar{\omega}_1 \tau} \quad (5)$$

$$\dot{n}_0 = 2\bar{\alpha}(iP_0 b_1^* e^{i\bar{\omega}_1 \tau} + c.c.) \quad (6)$$

where  $n_i = \langle \hat{n}_i \rangle$ ,  $b_i = \langle \hat{b}_i \rangle$ , and  $P_0 = \langle \hat{b}_0 \hat{b}_0 \rangle$ , and the dot now indicates a derivative with respect to the dimensionless time  $\tau = \kappa|\beta|^2 t$  with  $\bar{\alpha} = \alpha'/\kappa|\beta|^2$  and

$\bar{\omega}_1 = \omega'_1/\kappa|\beta|^2$ . We have also introduced a phenomenological decay rate,  $\bar{\Gamma}$ , for the intermediate state  $|1\rangle$  to describe losses due to inelastic collisions leading to vibrational decay for  $\omega_1 < 0$  or disintegration into pairs of atoms for  $\omega_1 > 0$ . Equations (2)-(6) are independent of the detuning  $\delta$ . This follows from the assumption that  $|\delta| \ll \kappa$  used to derive Eq. (1), which implies that the two-photon detuning is much less than the linewidth of the transition.

The equation of motion for the amplitude of the final state,  $b_3$ , can be immediately integrated to give

$$b_3(\tau) = \exp\left[\int_0^\tau d\tau'(-i\delta/\kappa|\beta|^2 + n_1(\tau'))\right]b_3(0). \quad (7)$$

Consequently, phase coherent amplification of  $|3\rangle$  only occurs when there is an initial signal to be amplified,  $b_3(0) \neq 0$ . Otherwise one only has amplification of the population as given by Eq. (2). This remains true even for the exact dynamics given by (1). For the remainder of this paper we let  $\delta = 0$ .

Consider first the case  $\bar{\omega}_1 = \bar{\Gamma} = 0$  where  $|1\rangle$  is initially in the vacuum state and  $n_3 \ll 1$ . Then for very short times such that the atomic condensate can be treated as an undepleted source with  $P_0(0) = n_0(0) = 2N$  the population in  $|3\rangle$  grows according to  $n_3(\tau) = \exp[2\bar{\alpha}^2(2N)^2\tau^3/3] - 1$ . This indicates that even for very short times the amplification cannot be described via a simple linear rate equation of the form  $dn_3/dt = Gn_3$  where  $G = \text{const}$ . For longer times,  $b_1$  and  $n_1$  both go to zero while  $n_3(\tau)$  saturates at a value that is less than its maximal value of  $n_3(0) + N$ , which implies less than 100% conversion of atoms into molecules (but approaching 100% for  $N \rightarrow \infty$ ). Fig. 1 shows the dynamics of the populations and amplitudes for the three modes. The population and amplitude in  $|3\rangle$  grow monotonically while states  $|1\rangle$  and  $|0\rangle$  initially oscillate at a frequency  $\sim \bar{\alpha}\sqrt{N}$ .

Figure 2 shows the coherent amplification of  $b_3$  and the amplification of the population  $n_3$ , which includes both coherent and incoherent contributions, for  $P_0(0) = n_0(0) = 2N = 500$ ,  $\bar{\alpha} = 1$ , and various values of  $\bar{\omega}_1$  and  $\bar{\Gamma}$ . As long as  $\bar{\omega}_1 \lesssim \bar{\alpha}\sqrt{N}$ , the  $|0\rangle \rightarrow |1\rangle$  transition is resonant and a finite  $\omega_1$  has no significant effect on the dynamics. For  $\bar{\omega}_1 \gg \bar{\alpha}\sqrt{N}$  the transition is off-resonant and this significantly slows the rate at which population builds up in  $|3\rangle$ . However, the value at which the population in  $|3\rangle$  saturates is unaffected by  $\bar{\omega}_1$ . Decay from the intermediate state only has a significant effect on the population and amplitude in  $|3\rangle$  for  $\bar{\Gamma}/2 \gtrsim 1 + n_3(0)$ . In this limit the decay out of  $|1\rangle$  dominates over the initial stimulated scattering from  $|1\rangle$  to  $|3\rangle$ . The fact that the dynamics are independent of  $\delta$  (for  $|\delta| \ll \kappa$ ) and only weakly dependent on  $\bar{\omega}_1$  indicate that mean-field energy shifts due to two-body interactions between atoms and molecules will have a negligible impact on the amplification of  $|3\rangle$ .

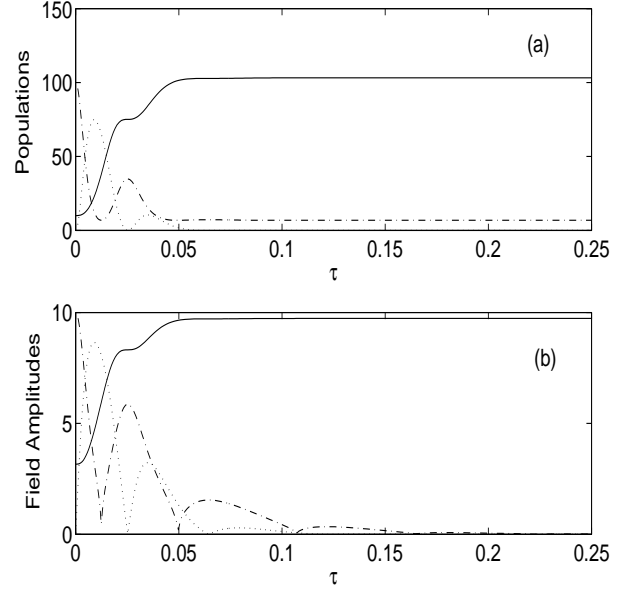


FIG. 1: (a) Populations of the three modes:  $n_3$  (solid line),  $n_1$  (dotted line), and  $n_0/2$  (dashed dot line). (b) Amplitudes of the three modes:  $|b_3|$  (solid line),  $|b_1|$  (dotted line),  $\sqrt{|P_0|/2}$  (dashed dot line). In both plots,  $\bar{\alpha} = 10$ ,  $\bar{\omega}_1 = \bar{\Gamma} = 0$ ,  $b_1(0) = n_1(0) = 0$ ,  $n_3(0) = b_3(0)^2 = 10$ , and  $n_0(0) = P_0(0) = 2N = 200$ .

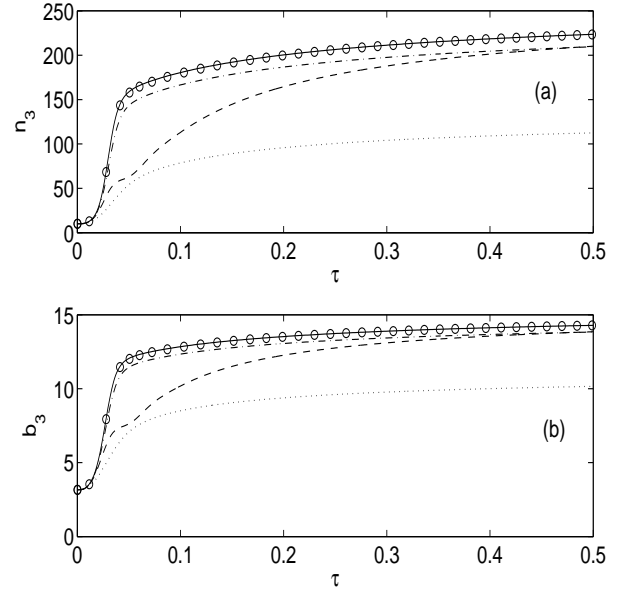


FIG. 2: (a) Population,  $n_3(\tau)$ , and (b) amplitude,  $b_3(\tau)$ , of the ground state molecular field for the initial conditions  $n_3(0) = b_3(0)^2 = 10$ ,  $n_0(0) = P_0(0) = 500$ , and  $n_1(0) = b_1(0) = 0$ . Solid line:  $\bar{\omega}_1/\bar{\alpha} = \bar{\Gamma} = 0$ ; Circles:  $\bar{\omega}_1/\bar{\alpha} = 10$  and  $\bar{\Gamma} = 0$ ; Dashed line:  $\bar{\omega}_1/\bar{\alpha} = 100$  and  $\bar{\Gamma} = 0$ ; Dashed dot line:  $\bar{\omega}_1/\bar{\alpha} = \bar{\Gamma} = 10$ ; Dotted line:  $\bar{\omega}_1/\bar{\alpha} = 10$  and  $\bar{\Gamma} = 100$ .

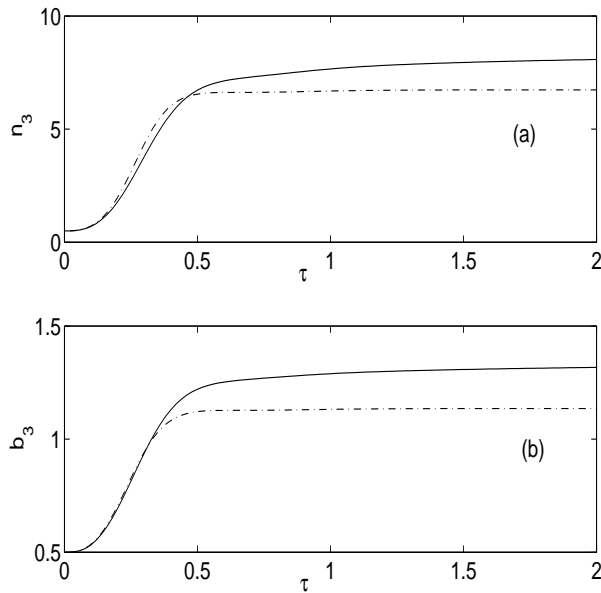


FIG. 3: (a) Population of the ground state molecular field,  $n_3(\tau)$ , calculated using Eq. (2) (dashed dot line) and directly from the master equation, Eq. (1), (solid line). (b) amplitude of ground state molecules,  $b_3(\tau)$ , from Eq. (7) (dashed dot line) and master equation (solid line). In both figures we have taken the initial amplitudes for state  $|3\rangle$  to be  $c_0 = c_1 = 1/\sqrt{2}$ ,  $\bar{\alpha} = 1$ ,  $\bar{\omega}_1 = \bar{\Gamma} = 0$ , and  $2N = 16$  to be the initial population in  $|0\rangle$  with  $|1\rangle$  initially in the vacuum state.

We can compare the solution obtained by integrating Eqs. (2)-(6) with the expectation values obtained from direct integration of the master equation, (1), for small numbers of molecules ( $\sim 10$ ). This is shown in Fig. 3. For Eq. (1) we choose the initial conditions to be a Fock state with occupation number  $2N$  for mode  $|0\rangle$ , and  $|3\rangle$  to be in an arbitrary superposition of 0 or 1 molecules,  $c_0|\text{vacuum}\rangle + c_1\hat{b}_3^\dagger|\text{vacuum}\rangle$ . For the factorized equations of motion we then have the corresponding initial conditions  $n_3(0) = |c_1|^2$ ,  $b_3(0) = c_0^*c_1$ , and  $n_0(0) = P_0(0) = 2N$ .

For short times, when the population and field amplitude in  $|3\rangle$  are growing exponentially, the two solutions show good agreement. For longer times,  $\tau \gtrsim 1/2$ , the two solutions for  $n_3$  and  $b_3$  saturate at different values. Numerical integration of Eq. (1) indicates that the conversion of atoms into ground state molecules asymptotically approaches 100% as  $\tau \rightarrow \infty$  for all  $N$  in contrast to the results obtained from Eqs. (2)-(6). This difference can be explained by noting that  $n_3(\tau)$  and  $b_3(\tau)$  saturate when no more new population forms in  $|1\rangle$ . From the equation of motion for  $\langle \hat{n}_1 \rangle$  one can see that the population in  $|1\rangle$  is driven by the cross-correlation  $\langle \hat{b}_1^\dagger \hat{b}_0 \hat{b}_0 \rangle$ . When this goes to zero, no more population is transferred from  $|0\rangle$  to  $|1\rangle$  and as a result  $n_3(\tau)$  saturates. Figure 4 shows  $\langle \hat{b}_1^\dagger \hat{b}_0 \hat{b}_0 \rangle$  calculated directly from the master equation and  $b_1^*P_0$  calculated using Eqs. (2)-(6). As one can see,  $b_1^*P_0$  decays to zero much faster than  $\langle \hat{b}_1^\dagger \hat{b}_0 \hat{b}_0 \rangle$ . Con-

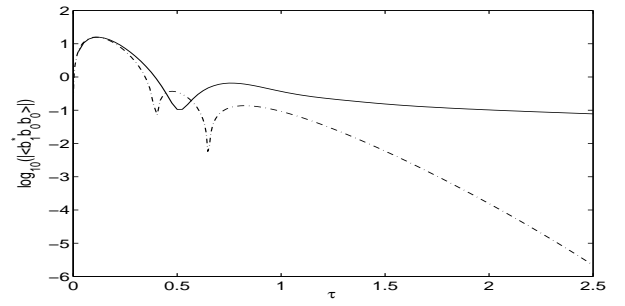


FIG. 4:  $\langle \hat{b}_1^\dagger \hat{b}_0 \hat{b}_0 \rangle$  calculated from the master equation (solid line) and  $b_1^*P_0$  calculated from factorized equations of motion (dashed dot line). All parameters are the same as Fig. 3.

sequently the buildup of population in  $|1\rangle$  as given by Eq. (4) stops well before that given by the exact quantum dynamics. We note that other cross-correlations involving the intermediate state such as  $\langle \hat{n}_3 \hat{n}_1 \rangle$  exhibit similar behavior with the uncorrelated factorized values obtained from Eqs. (2)-(6) decaying to zero much faster than the exact values obtained from the master equation.

In conclusion we have analyzed a model for a coherent matter-wave amplifier of ground-state molecules in an optical cavity by analyzing semiclassical rate equations and comparing them to the exact quantum dynamics.

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